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Femtosecond shock wave dynamics of insensitive energetic materials

5. FUNDING NUMBERS  
F49620-97-1-0056

6. AUTHOR(S)  
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REPORT NUMBER

9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES)  
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## 13. ABSTRACT (Maximum 200 Words)

The goal of this project is to develop a fundamental understanding, at the molecular level, of the initial events occurring during the first picoseconds of impact initiation of energetic materials. It is believed this fundamental research will lead to a clearer picture of the factors which determine the sensitivity of energetic materials to shock impact initiation. Understanding and perhaps controlling sensitivity will result in safer explosives. In a larger sense, the problem of molecular level understanding of chemical and physical phenomena involved in impact processes has much wider and more general implications for such relevant processes as mechanical failure, crack propagation, lubrication and wear, etc. At the present time experimental techniques to study these problems are lacking. By developing such techniques with the goal of understanding impact initiation of energetic materials, these other areas may be addressed as well.

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### 2-3. Status of Effort (Objectives have not changed)

The goal of this project is to develop a fundamental understanding, at the molecular level, of the initial events occurring during the first picoseconds of impact initiation of energetic materials. It is believed this fundamental research will lead to a clearer picture of the factors which determine the sensitivity of energetic materials to shock impact initiation. Understanding and perhaps controlling sensitivity will result in safer explosives. In a larger sense, the problem of molecular level understanding of chemical and physical phenomena involved in impact processes has much wider and more general implications for such relevant processes as mechanical failure, crack propagation, lubrication and wear, etc. At the present time experimental techniques to study these problems are lacking. By developing such techniques with the goal of understanding impact initiation of energetic materials, these other areas may be addressed as well.

There are three main experimental set-ups used in this work: (1) the nanoshock set-up, which uses a picosecond laser to pump and probe shock waves in molecular materials with absorption spectroscopy or coherent anti-Stokes Raman (CARS); (2) the surface set-up, which uses a femtosecond laser to pump and probe shock waves at surfaces, and (3) the energy transfer set up, which uses a femtosecond infrared laser to study molecular mechanical energy transfer processes in condensed phases.

With the nanoshock set up, we developed a method for generating powerful (up to 5 GPa) and reproducible shock waves with a fast rise time  $<25$  ps, at a high repetition rate of 100/s. This versatile tool has been used to study anthracene, a model system, NTO an insensitive energetic material, polymers such as PMMA, and pore collapse that leads to hot spot formation.

In the energy transfer work, we use an infrared laser to pump a molecular vibration (e.g. C-H or OH stretch) and a visible pulse to probe where the energy is going using anti-Stokes Raman scattering (2-D vibrational spectroscopy). The motivation is to understand the roles and pathways of vibrational energy transfer in shock initiation. This is accomplished by studying energetic materials and nonenergetic materials. The reason for studying nonenergetic materials is to build up a fundamental understanding of vibrational energy flow and to see how energetic materials differ from ordinary materials. We have completed extensive studies of nitromethane, acetonitrile, methanol, benzene, and water. Through a program of continuous improvement and technique development, we have discovered how to use this system to perform "three dimensional vibrational spectroscopy" measurements. The 3D technique is presently the most powerful method for understanding vibrational energy flow in condensed phases, which plays a pivotal role in essentially all chemical processes.

The surface set-up involves ultrafast vibrational sum-frequency generation (SFG) measurements. SFG is a powerful technique that involves mixing a vibrational IR pulse and a visible pulse at a surface or interface. SFG solves two critical problems simultaneously: even though there are not very many molecules on a surface, the coherent SFG signal is strong enough to be readily detected; even though there are many more molecules in the bulk than at a surface or interface, SFG occurs only from the interfacial molecules. The apparatus to perform ultrafast SFG experiments is quite complicated and it took a long time to construct and test. Finally we

have what is arguably the most powerful and sensitive SFG system ever. This is made possible by the high power of the Ti:sapphire laser, which has two amplification stages, and our IR OPA which is highly efficient. Preliminary SFG spectra are being obtained. This system has many applications. First the time resolution obtained in a shock experiment is (when all other problems are solved) limited by the thickness of the shocked material. By having this thickness a monolayer, we should obtain ultimate resolution. With nanoshocks, we achieved ~50 ps time resolution. With the surface apparatus, we hope to get ~1 ps time resolution. Second, it is very interesting to look at shocked materials with a surface-selective probe. The applications are crack generation and propagation, spallation, and dynamic lubrication and wear at moving interfaces. For example, in the problem of the spallation of a ceramic coating from a metal plate, which can be induced by a laser-driven shock, these techniques will focus only on the metal-ceramic interface, where the action really is, rather than in the bulk ceramic or metal.

#### 4. Accomplishments/new findings

- High power picosecond mid-IR optical parametric amplifier for infrared-Raman spectroscopy
- First ultrafast vibrational spectroscopy of shock waves in high explosives
- First detailed study of vibrational energy transfer in a high explosive (nitromethane)
- Real time spectroscopic observation of mechanical relaxation in shocked polymers
- Real time spectroscopic observation of effects of pore collapse leading to hot spot generation
- 3D vibrational spectroscopy of molecular liquids
- Third order nonlinear optical properties of sulfur-rich compounds
- Ultrafast measurements of shear relaxation in shock-compressed polymers

#### 5. Personnel Supported

##### Number of Contract/Grant Co-Investigators\*

Faculty	1	Post Doctorates	3	Graduate Students	2	Other	0
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1. Dana D. Dlott, PI, leads the project.
2. Dr. John Deak, a postdoctoral associate, performed studies of energy transfer in high explosives and then took a position at Proctor and Gamble, Inc., in Nov. 1999.
3. Mr. Larry Iwaki, a graduate student, worked on the energy transfer project, obtained his Ph.D. degree in Jan. 2000 and took a postdoctoral position at NIST
4. Mr. Jens Franken, a graduate student, obtained his Ph.D. degree in Jan. 1999 and took a postdoctoral position at the Institute of Atomic and Molecular Sciences in Taiwan..
5. Dr. Selezion Hambir, a postdoc, is working on the ps CARS project.
6. Dr. J. J. Cavaleri, a postdoctoral associate, worked on shock wave development. He left in 1999 to take a permanent position with the Los Angeles Sheriff's Office.

## 6. Publications Related to Aforementioned Contract/Grant

1. Name of Journal: Phys. Rev. Lett.

Title of Article: Ultrafast Raman spectroscopy of shock fronts in molecular solids

Authors: Guray Tas, Jens Franken, Selezion A. Hambir and Dana D. Dlott

Volume 78                      Pages pp. 4585-4588                      Month Published: June                      Year: 1997

2. Name of Journal: J. Appl. Phys

Title of Article: Coherent Raman spectroscopy of nanoshocks

Authors: Guray Tas, Selezion A. Hambir, Jens Franken, David E. Hare and Dana D. Dlott

Volume 82                      Pages 1080-1087                      Month Published: Aug.                      Year: 1997

3. Name of Journal: *Shock Compression of Condensed Matter--1997*, Stephen Schmidt et. al., eds., AIP conference proceedings vol. 429.

Title of Article: Ultrafast vibrational spectroscopy of shocks in molecular materials: the first 100 ps

Authors: Selezion A. Hambir, Jens Franken, Jeffrey R. Hill and Dana D. Dlott

Volume 429                      Pages 823-826                      Month Published: xxx                      Year: 1998

4. Name of Journal: *Shock Compression of Condensed Matter--1997*, Stephen Schmidt et. al., eds. AIP conference proceedings vol. 429.

Title of Article: Picosecond vibrational spectroscopy of shocked energetic materials

Authors: Jens Franken, Selezion A. Hambir and Dana D. Dlott

Volume 429                      Pages 819-822                      Month Published: xxx                      Year: 1998

5. Name of Journal: Opt. Lett

Title of Article: High power picosecond mid-infrared optical parametric amplifier for infrared-Raman spectroscopy

Authors: John C. Deak, Lawrence K. Iwaki and Dana D. Dlott

Volume 22                      Pages 1796-1798                      Month Published: December Year: 1997

6. Name of Journal: Rev. High Press. Sci. Tech.

Title of Article: Ultrafast Spectroscopy of Laser-driven Nanoshocks in Molecular Crystals

Authors: Selezion A. Hambir, Jens Franken and Dana D. Dlott

Volume 7                      Pages 891-896                      Month Published: Aug.                      Year: 1998

7. Name of Journal: J. Phys. Chem. B.

Title of Article: The new wave in shock waves

Authors: Dana D. Dlott, Selezion Hambir and Jens Franken

Volume 102                      Pages 2121-2130                      Month Published:                      Year: 1998

8. Name of Journal: J. Appl. Phys.

Title of Article: Orientation dependence of shock-induced heating in anharmonic molecular crystals

Authors: V. K. Jindal and Dana D. Dlott

Volume 83                      Pages 5203-5211                      Month Published: May                      Year: 1998

9. Name of Journal: Mol. Cryst. Liq. Cryst.

Title of Article: Ultrafast Spectroscopy of Laser-driven Shock Waves in Molecular Materials

Authors: Jens Franken, Selezion A. Hambir and Dana D. Dlott

Volume 314                      Pages 25-36                      Month Published: xxx                      Year: 1998

10. Name of Journal: Chem. Phys. Lett.

Title of Article: When Vibrations Interact: Ultrafast Energy Relaxation of Vibrational Pairs in Polyatomic Liquids

Authors: John C. Deak, Lawrence K. Iwaki and Dana D. Dlott

Volume 293                      Pages 405-411                      Month Published: Sept.                      Year: 1998

11. Name of Journal: J. Phys. Chem. A

Title of Article: Vibrational Energy Relaxation of Polyatomic Molecules in Liquids: Acetonitrile

Authors: John C. Deak, Lawrence K. Iwaki and Dana D. Dlott

Volume 102                      Pages 8193-8201                      Month Published: Sept.                      Year: 1998

12. Name of Journal: Ph.D. Dissertation

Title: Picosecond vibrational spectroscopy of shocked solids

Authors: Jens Franken

Volume xxx                      Pages xxx                      Month Published: Jan                      Year: 1999

13. Name of Journal: J. Appl. Phys.

Title of Article: Ultrafast shock-induced orientation of polycrystalline films: applications to high explosives

Authors: Jens Franken, Selezion A. Hambir and Dana D. Dlott

Volume 85                      Pages 2068-2074                      Month Published: Feb.                      Year: 1999

14. Name of Journal: J. Phys. Chem. A

Title of Article: Vibrational Energy Redistribution in Polyatomic Liquids: Ultrafast IR-Raman Spectroscopy of Nitromethane

Authors: " John C. Deak, Lawrence K. Iwaki and Dana D. Dlott

Volume 103                      Pages 971-979                      Month Published: Feb.                      Year: 1999

15. Name of Journal: Chem. Phys. Lett.

Title of Article: Vibrational energy redistribution in liquid benzene

Authors: Lawrence K. Iwaki, John C. Deak, Stuart T. Rhea and Dana D. Dlott

Volume 303                      Pages 176-182                      Month Published: Apr.                      Year: 1999

16. Name of Journal: J. Phys. Chem. A.

Title of Article: Third order nonlinear optical properties of sulfur-rich compounds

Authors: Jonathan G. Breitzer, Dana D. Dlott, Lawrence K. Iwaki, Sean M. Kirkpatrick and Thomas B. Rauchfuss

Volume 103            Pages 6930-6937            Month Published: Aug.            Year: 1999

17. Name of Journal: Annu. Rev. Phys. Chem.

Title of Article: Ultrafast spectroscopy of shock waves in molecular materials

Authors: Dana D. Dlott

Volume 50            Pages 251-78            Month Published: Dec.            Year: 1999

18. Name of Journal: Shock Compression of Condensed Matter--1999

Title of Article: Ultrafast dynamics of nanoshocks in molecular materials

Authors: S. A. Hambir, H. Kim and Dana D. Dlott

Volume 505            Pages 945-950            Month Published: Sept.            Year: 2000

19. Name of Journal: Acc. Chem. Res.

Title of Article: Nanoshocks in molecular materials

Authors: Dana D. Dlott

Volume 33            Pages 37-45            Month Published: Jan.            Year: 2000

20. Name of Journal: J. Raman. Spectr.

Title of Article: Ultrafast Infrared-Raman Studies of Vibrational Energy Redistribution in Polyatomic Liquids"

Authors: John C. Deak, Lawrence K. Iwaki, Stuart T. Rhea, and Dana D. Dlott

Volume 31            Pages 263-274            Month Published: Apr.            Year: 2000

21. Name of Book: Ultrafast Infrared and Raman Spectroscopy", M. D. Fayer, ed (in press).

Title of Article: Vibrational Energy Redistribution in Polyatomic Liquids: Ultrafast IR-Raman Spectroscopy

Authors: Lawrence K. Iwaki, John C. Deak, Stuart T. Rhea and Dana D. Dlott

Volume xxx            Pages xxx            Month Published: xxxx.            Year: 2000

22. Name of Journal: Ph.D. Dissertation



Title of Article: Vibrational energy relaxation in liquids

Authors: Lawrence K. Iwaki, Jr.

Volume                      Pages 129                      Month Published: Jan.                      Year: 2000

23. Name of Journal: Phys. Rev. Lett.

Title of Article: Ultrafast dynamics of shock waves in polymers and proteins: the energy landscape

Authors: Hackjin Kim, Selezione A. Hambir and Dana D. Dlott

Volume 83                      Pages 5034-5037                      Month Published: Dec.                      Year: 1999

24. Name of Journal: J. Phys. Chem. A

Title of Article: Shock compression of organic polymers and proteins: ultrafast structural relaxation dynamics and energy landscapes

Authors: Hackjin Kim, Selezione A. Hambir and Dana D. Dlott

Volume 104                      Pages 4239-4252                      Month Published: Mar.                      Year: 2000

25. Name of Journal: J. Phys. Chem. A

Title of Article: Vibrational energy relaxation in liquid water and deuterated water

Authors: John C. Deak, Stuart T. Rhea, Lawrence K. Iwaki and Dana D. Dlott

Volume 104                      Pages 4866-4875                      Month Published: June                      Year: 2000

26. Name of Journal: Chem. Phys. Lett.

Title of Article: Ultrafast vibrational energy redistribution among C-H and O-H stretching modes of liquid methanol

Authors: Lawrence K. Iwaki and Dana D. Dlott

Volume 41                      Pages 419-425                      Month Published: May                      Year: 2000

27. Name of Journal: J. Phys. Chem. A

Title of Article: Three-dimensional spectroscopy of vibrational energy relaxation in liquid methanol

Authors: Lawrence K. Iwaki and Dana D. Dlott

Volume 104

Pages 9101-9112

Month Published: Oct.

Year: 2000

28. Name of Book: Proceedings of the High Energy Density Matter Contractors Conference

Title of Article: Ultrafast dynamics of energetic materials: understanding impact sensitivity

Authors: Dana D. Dlott

Volume

Pages 100-104

Month Published: Sept.

Year: 2000

## 7. Interactions/Transitions

### a. Conferences attended and presentations made

a1. (*invited*) National Institute for Research in Inorganic Materials, Tsukuba, Japan (Jan., '97), "Shock waves in molecular solids: Ultrafast vibrational spectroscopy of the first nanosecond".

a2. (*invited*) Kyushu University, Fukuoka, Japan (Jan. '97), "Ultrafast spectroscopy of shock waves in solids".

a3. (*invited*) Kyushu University, Fukuoka, Japan (Jan. '97), "Vibrational echo studies of heme protein dynamics".

a4. (*invited*) Osaka University, Osaka, Japan (Jan. '97), "Ultrafast spectroscopy of shock waves in solids".

a5. (*invited*) American Physical Society National Meeting, Kansas City, MO (Mar. '97), "Vibrational echo studies of protein dynamics".

a6. (*invited*) 50th Annual Meeting of the Society for Imaging Science and Technology, Cambridge, MA (May. '97), "Pulse duration dependence for laser photothermal imaging media".

a7. (*invited*) Abbott Laboratories, North Chicago, IL (May '97) "Ultrafast imaging with near infrared pulses".

a8. (*invited*) US Army Research Office, Research Triangle Park, NC, Annual Chemistry Program Review (June '97), "Ultrashort light pulses and energetic materials".

a9. (*invited*) Eighth International Conference on Unconventional Photoactive Systems, Nara, Japan (Aug. '97), "Ultrafast dynamics of shock waves in molecular materials".

- a10. (*invited*) Joint AIRAPT-16 and HPCJ-38 International Conference on High Pressure Science and Technology, Kyoto, Japan (Aug. '97) "Ultrafast dynamics of shock waves in molecular materials".
- a11. (*invited*) SSSW'97 Summer School on Shock Waves, 1997, Lake Biwa, Japan (Aug. '97), "The new wave in shock waves".
- a12. (*invited*) University of Illinois, Department of Chemistry, (Sept. '97), "The new wave in shock waves".
- a13. (*invited*) Pennsylvania State University, Department of Chemistry, (Oct. '97), "Ultrafast dynamics of shock waves in molecular materials".
- a14. (*invited*) American Physical Society, Southeastern Regional Meeting (Nov. '97), "Ultrafast dynamics of shock waves in molecular materials".
- a15. (*invited*) Los Alamos National Laboratory (Nov. '97), "Vibrational echo and shock wave studies of heme protein dynamics".
- a16. (*invited*) Los Alamos National Laboratory (Nov. '97), "Ultrafast dynamics of shock waves in molecular materials".
- a17. (*invited*) Kansas State University (Dec. '97), "Ultrafast spectroscopy of molecular materials: the new wave"
- a18. (*invited*) Markem Corp., Keene, NH (Dec. '97) "Ultrafast imaging using near infrared lasers".
- a19. (*invited*) Office of Naval Research Workshop on Energetic Materials (Jan. '98), "Ultrafast dynamics of energetic materials".
- a20. (*invited*) Department of Physics, Washington State University, Pullman, WA (May '98), "Ultrafast spectroscopy of shock waves in molecular materials".
- a21. (*invited*) Department of Chemistry, University of Washington, Seattle, WA (May '98), "Ultrafast infrared spectroscopy of vibrational relaxation in proteins and simple liquids"
- a22. (*invited*) Department of Chemistry and Geology, University of Washington, Seattle, WA (May '98), "Ultrafast vibrational spectroscopy of shock waves in molecular materials".
- a23. (*invited*) 1998 Air Force Office of Scientific Research Molecular Dynamics Conference, Monterey, CA (May '98), "Ultrafast vibrational dynamics in condensed phases".
- a24. (*invited*) American Chemical Society, National Meeting, Boston, MA (Aug. '98) "Ultrafast Vibrational Echo and Shock Wave Studies of Heme Protein Dynamics".

- a25. (*invited*) Department of Chemistry, Colorado State University, Ft. Collins, CO (Sept. '98), "Vibrational energy transfer in polyatomic liquids".
- a26. (*invited*) Department of Chemistry, University of Colorado, Boulder, CO (Sept. '98), "Vibrational energy transfer in polyatomic liquids".
- a27. (*invited*) Southeast Regional Meeting of American Physical Society, Miami, FL (Nov. '98), "Ultrafast spectroscopy of shock waves in biophysics".
- a28. (*invited*) Fifth International Conference on Molecular Dynamics in Condensed Phase, Newport Beach, CA (Feb. '99), "Vibrational energy relaxation in polyatomic liquids".
- a29. (*invited*) DOE Basic Energy Sciences workshop on High-Pressure Research, Washington, D. C. (June '99) "New frontiers for high pressure: fast dynamics and large molecules".
- a30. (*invited*) 1999 High Energy Density Matter AFOSR Contractor's Meeting, Cocoa Beach, FL (June '99), "Ultrafast dynamics of energetic materials: towards an understanding of impact sensitivity"
- a31. (*invited*) American Physical Society topical conference on Shock Compression in Condensed Matter, Snowbird Utah (June '99). "Ultrafast dynamics of nanoshocks in molecular materials".
- a32. (*invited*) International workshop on new models and predictive methods for shock wave/dynamic processes in energetic materials and related solids, University of Maryland, College Park, MD (July '99), "Ultrafast dynamics of energetic materials: towards an understanding of impact sensitivity".
- a33. (*invited*) Protein dynamics workshop, Telluride, CO (July '99). "Shock wave studies of protein dynamics".
- a34. (*invited*) American Chemical Society National Meeting, New Orleans, LA (Aug. '99), "Ultrafast two-dimensional vibrational spectroscopy of water".
- a35. (*invited*) International Laser Science meeting, Santa Clara, CA (Oct. '99), "Ultrafast two-dimensional vibrational spectroscopy of water".
- a36. (*invited*) Ohio State University, Department of Chemistry, Columbus, OH (Oct. '99), "Two-dimensional vibrational spectroscopy of liquids".
- a37. (*invited*) Beckman Laser Institute, University of California, Irvine, CA (Oct. '99), "Ultrafast spectroscopy of nanoshocks in biological materials".
- a38. (*invited*) University of California, Department of Chemistry, Irvine, CA, (Oct. '99) "Two-dimensional vibrational spectroscopy of liquids".

a39. (*invited*) Wellman Laboratories of Photomedicine, Massachusetts General Hospital, Boston, MA (Oct. '99), "Ultrafast spectroscopy of nanoshocks in biological materials".

**b. Consultative and advisory functions**

b.1. I am a member of the nominations committee for the American Physical Society Topical Group on Shock Compression of Condensed Matter. That committee comes up with the name of people who agree to do all the work running the biennial conference, writing the newsletters, etc.

b.2. With Dr. Bob Morris of AFRL, I organized a symposium for the Aug. 2000 national meeting of the ACS, titled, "Chemistry in extreme conditions".

b.3. I was appointed as an advisory editor for Applied Physics Letters and the Journal of Applied Physics, from 2000-2002

b.4. I am on the program committee for the APS International Conference on Shock Compression in Condensed Matter

b.5. I am organizing the Sixth International Conference on Molecular Reaction Dynamics in Condensed Matter

**c. Transitions**

c.1. Researchers at Abbott Labs (North Chicago, IL) are investigating the medical applications of laser driven shock waves. I consulted with them earlier this year on how to optimize the efficiency of their methods using the laser shock wave methods developed here. Contact: Dr. Mark R. Pope, 847-937-1171.

c.2. I discussed ways of using laser driven shock waves to print security codes on expensive integrated circuit packages with Markem, Corp. Contact: Dr. Alan Boyer, (603) 352-1130.

c.3. Researchers at Los Alamos National Labs are evaluating our molecular thermometry techniques for possible application to energetic materials subjected to impact, and I had extensive discussions and reviewed their report titled, "Molecular Thermometry of Energetic Materials Subjected to Impact". Contact: Dr. Brian Fishbine, 505-667-3583.

**d. New discoveries, inventions or patent disclosures**

None

**9. Honors and Awards**

Fellow of the American Physical Society, 1997 (Dlott)  
Fellow of the Optical Society of America, 1999 (Dlott)  
Associate, Center for Advanced Study, 1999 (Dlott)